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AUTORADIOGRAPHIC TECHNIQUE FOR RAPID INVENTORY OF PLUTONIUM-CONTAINING FAST CRITICAL ASSEMBLY FUEL

by

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Nondestructive Assay Section
Special Materials Division

October 1977

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AUTORADIOGRAPHIC TECHNIQUE FOR RAPID INVENTORY OF PLUTONIUM-CONTAINING FAST CRITICAL ASSEMBLY FUEL

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ABSTRACT

A nondestructive autoradiographic technique is described which can provide a verification of the piece count and the plutonium content of plutonium-containing fuel elements. This technique uses the spontaneously emitted gamma rays from plutonium to form images of fuel elements on photographic film. Autoradiography has the advantage of providing an inventory verification without the opening of containers or the handling of fuel elements. Missing fuel elements, substitution of nonradioactive material, and substitution of elements of different size are detectable. Results are presented for fuel elements in various storage configurations and for fuel elements contained in a fast critical assembly.

I. INTRODUCTION

Increased safeguards, such as those proposed for ERDA Manual Chapter 7401, will require more frequent verification of special nuclear material (SNM) inventories. Needed are inventory techniques which can verify the piece count and the SNM content of large populations of fuel elements, and which can minimize inventory time, radiation exposure of personnel, and interference with normal operations. Examples of facilities where large numbers of SNM-containing fuel elements require improved inventory techniques are the fast critical assemblies operated by the Argonne National Laboratory.

Previous inventory techniques applied to Argonne fast critical assembly fuel were time-consuming and disruptive, and resulted in large radiation doses to personnel. Fuel elements were counted visually and were sampled manually for nondestructive assay. Work rules for plutonium required that all fuel storage containers be opened in special hoods outside the storage vaults and be examined by health-physics personnel. Criticality rules required that only a few storage containers be examined at one time. This procedure prevented normal reactor loading change operations during the inventory. Recently, autoradiographic techniques have been developed for inventories of plutonium-containing fuel at the Argonne fast critical assemblies.

Autoradiography has been used for many years to study the distribution of fissile material and fission products in reactor fuel.[1-4] The spontaneously emitted X- and gamma-radiation from plutonium-containing fuel elements exposes photographic film. The 60-keV gamma ray from Am-241 is the most intense radiation from Argonne fast critical-assembly fuel. This radiation can produce an image of a source fuel element on film which is in contact with a fuel-storage container. Counting the images on the film then gives a piece count of fuel elements without having to open the container or handle the fuel elements. For fixed exposure time, isotopic composition, absorber, and geometry, the image density produced by an isolated fuel element is proportional to the concentration of plutonium in that element. This allows a simultaneous attribute check to be made for plutonium content. The technique is a qualitative, or a semiquantitative rather than a quantitative indication of plutonium concentration.

The following sections will describe the autoradiographic inventory technique in detail and will show results obtained with plutonium-containing fuel elements in various containment modes.

II. FUEL IN STORAGE

A. Plates

A large part of the Argonne fast critical-assembly fuel is in the form of stainless steel-clad plutonium-alloy plates. A large fraction of these plates are 2" wide, 1/4" thick, and from 1" to 8" long, with 0.015"-thick cladding. A small fraction of the plates are 1/8" thick (1" = 2.54 cm).

1. Argonne-East Storage

Fuel plates at the Argonne-Illinois (Argonne-East) site are stored in cylindrical canisters made of 0.213" thick aluminum. These canisters are approximately 6.5" in diameter and 9" high. Each contains an insert, made of 0.018"-thick stainless steel, which holds the plates in place. The plates are inserted lengthwise into the holder, with the 1/4" edge toward the cylinder wall. This container is shown in Fig. 1. The canister will accommodate 18 plates, 5" to 8" long. More than eighteen 4" or shorter plates can be stored by forming more than one layer.

Autoradiographs of fuel plates were obtained by wrapping strips of prepackaged film around the outside of the canister. The film used for these and all other autoradiographs in this report was Kodak-AA industrial x-ray film. All film was developed in a Kodak X-OMAT Model B automatic processor. The exposure time was typically 45 minutes for plutonium containing 11.5% isotope 240, and was shorter for plutonium higher in isotope 240. A lead letter was attached to the canister under the film so that the point of overlap could be identified. An autoradiograph of 18 plates in a canister is shown in Fig. 2. The lower half of this autoradiograph is lighter than the upper half because of gamma-ray absorption by the stainless steel insert, which ends in the center of the autoradiograph.

With this technique, missing fuel elements and the substitution of nonradioactive material can both be readily detected. An example of a missing element is illustrated in Fig. 3A. Here, the center element is missing, and there are four plutonium-containing plates on either side of the void. The image of the stainless steel insert, visible at the top of this autoradiograph, was caused by radiation from neighboring plates being scattered by the insert.

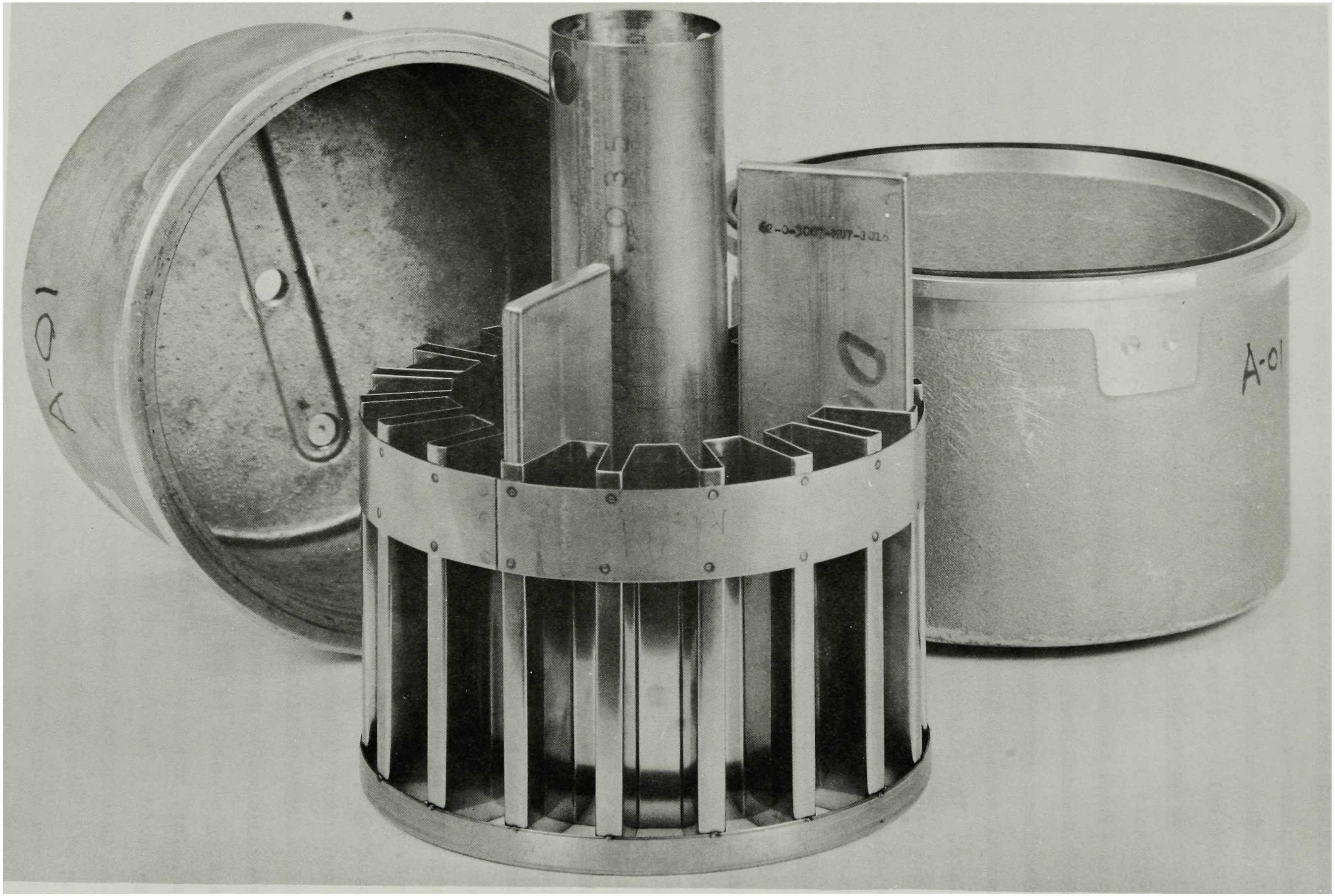

Fig. 1. Argonne-East fuel plate storage canister.
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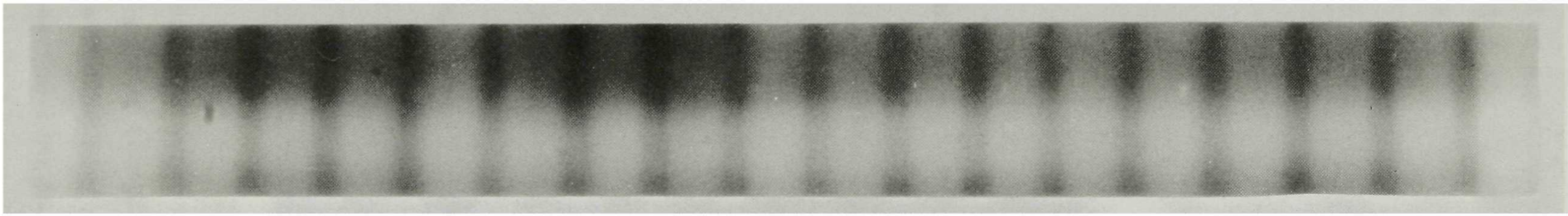

Fig. 2. Autoradiograph of 18 plutonium-containing plates in an Argonne-East canister. ANL Neg. No. 150-77-13.

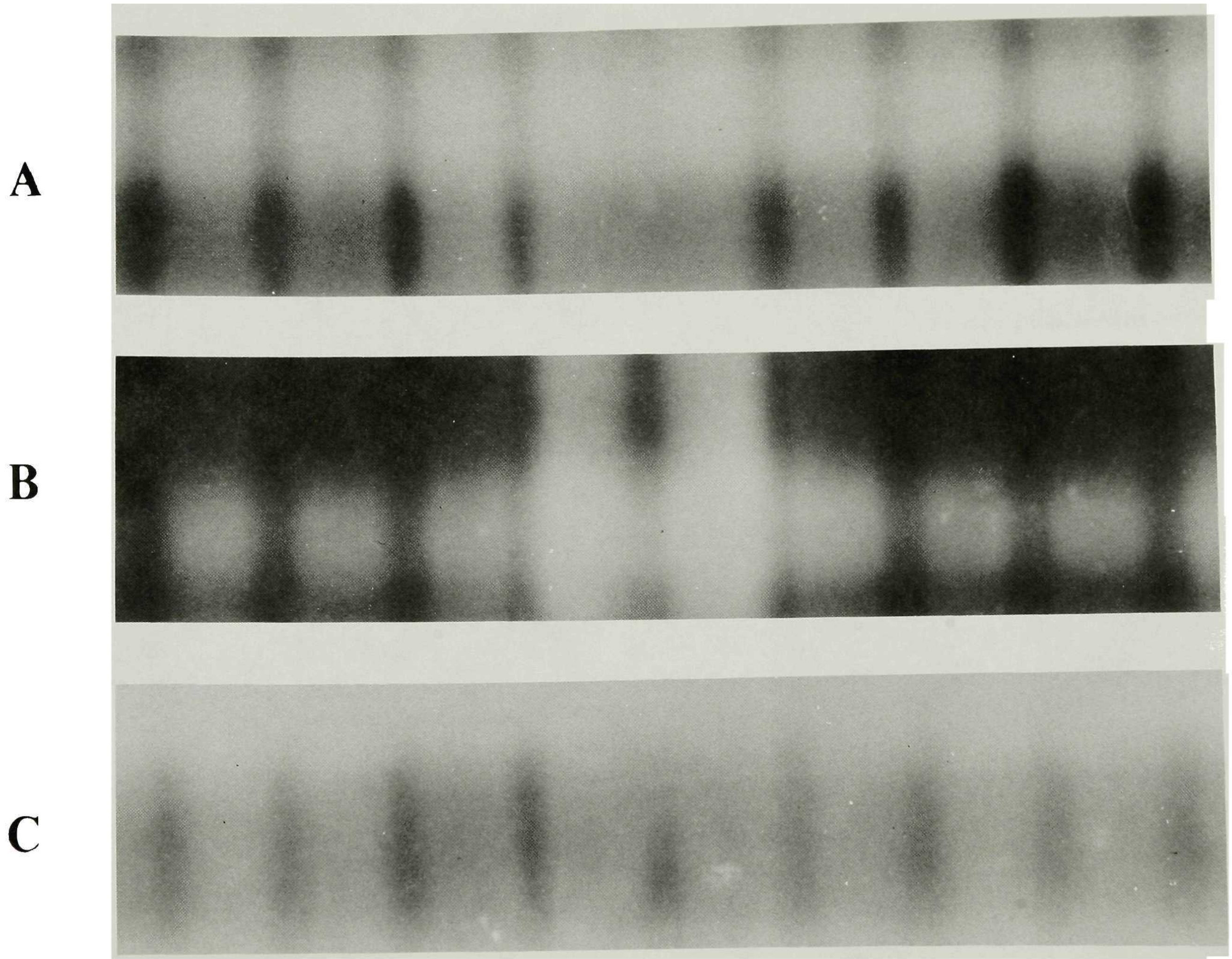

Fig. 3. Segments of autoradiographs of fuel plates in Argonne-East canisters
A. Center plate is missing
B. Center plate is solid stainless steel
C. Center plate is one inch shorter than its neighbors
ANL Neg. No. 150-77-12.

Figure 3B is an example of the results obtained when a nonradioactive element is substituted for a plutonium-containing plate. Here, the center plate is made of solid stainless steel with the same dimensions as a plutonium-containing plate. The steel plate appears to act as a scatterer of radiation from the neighboring plates, with the scattered radiation forming an image similar to that obtained for a plutonium-containing plate. However, the steel-plate image and the area surrounding the image are lighter, because the steel plate contributes no direct radiation and no radiation to be scattered by its neighbors. As there is less radiation to be scattered, the neighboring plutonium-containing plates also have less dense images.

The autoradiographic technique can also be used to verify the length of the plates in a storage container. If the film is placed around the canister so that the tops of the plates are visible, a shorter or a longer plate can be detected. An example of a short plate is shown in Fig. 3C. Here, the central plate is one inch shorter than its neighbors. Normally, only plates of one length are stored in a given container, and the length is recorded on a tag for each container. In Fig. 3, the autoradiographs were made with 7.0-cm wide film and exposed for approximately 45 minutes.

A full-scale inventory of 3,064 plates in 172 canisters at Argonne-East was performed with the use of the autoradiographic technique. The inventory was successfully completed with a saving in time and in radiation exposure, and with minimal impact on normal operations.

2. Argonne-West Storage

Fuel plates at the Argonne-Idaho (Argonne-West) site are stored in flat, rectangular aluminum canisters. Top and bottom walls are 0.216" thick, and the inside dimensions are approximately 2" x 6" x 8". An array of posts inside the canisters allows the plates to be stored on their 1/4" edge. A row of 12 plates can be stored across the width of the canister, or a row of 16 across the length. More than sixteen 4" or shorter plates can be accommodated by putting more than one plate in each row. Work rules require that all plates in any one canister must be the same size. The canister is illustrated in Fig. 4.

In the storage vault, each canister resides in its own cubicle. Autoradiographs of the canister contents are obtained by inserting a film packet under the canister in its cubicle, exposing the film for about 45 minutes, and then removing the packet. A typical autoradiograph of a canister filled to capacity with twenty-four 4"-long plates is shown in Fig. 5. As in Fig. 3, the density of each plate image has a direct radiation component and a scattered radiation component. Each plate can scatter radiation from its neighbors. Because plates at the edge of an array have only one neighbor, their scattered radiation is less, and their images are less dense than those of plates with two neighbors. This effect can be seen in Fig. 5.

A missing element in an array of plates is easily detected, as seen in Fig. 6, which shows an autoradiograph of twelve 4"-long plates, with 7 plates on one side and 5 on the other. There is a vacant position between the third and fourth plates on the side with 5 plates.

Substitution of nonradioactive material for a plutonium-containing fuel element is also detectable, as illustrated in Fig. 7. This shows an array of 14 plates: 13 plutonium-containing plates and 1 stainless steel plate in the center of one row. The image density of the steel plate is believed to be due exclusively to scattering of radiation from its neighbors. Because there is no direct radiation contribution by the steel plate, its image is less dense than is the image of a plutonium-containing plate with two plutonium-containing neighbors. The two plutonium-containing neighbors of the steel plate have only one plutonium-containing neighbor themselves. As a result, less radiation is scattered, and their image density is reduced to approximately that of the corner plates. This is expected, as the corner plates and the plates next to the steel element are all plutonium-containing plates with only one plutonium-containing neighbor. The substitution of such materials as depleted uranium or lead gives essentially the same results as seen in Fig. 7. If a less dense material, such as aluminum, is substituted, the

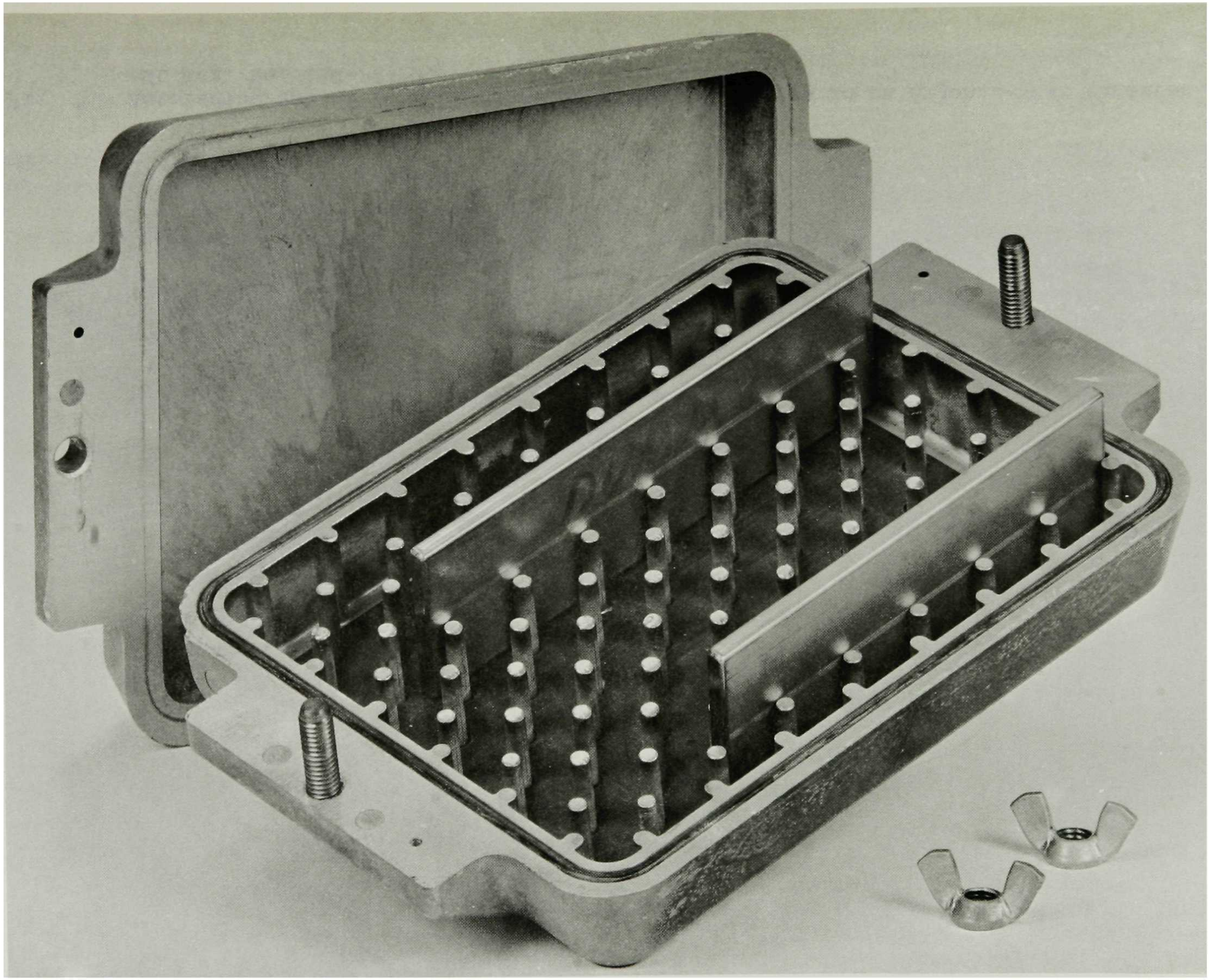

Fig. 4. Argonne-West fuel plate storage canister.
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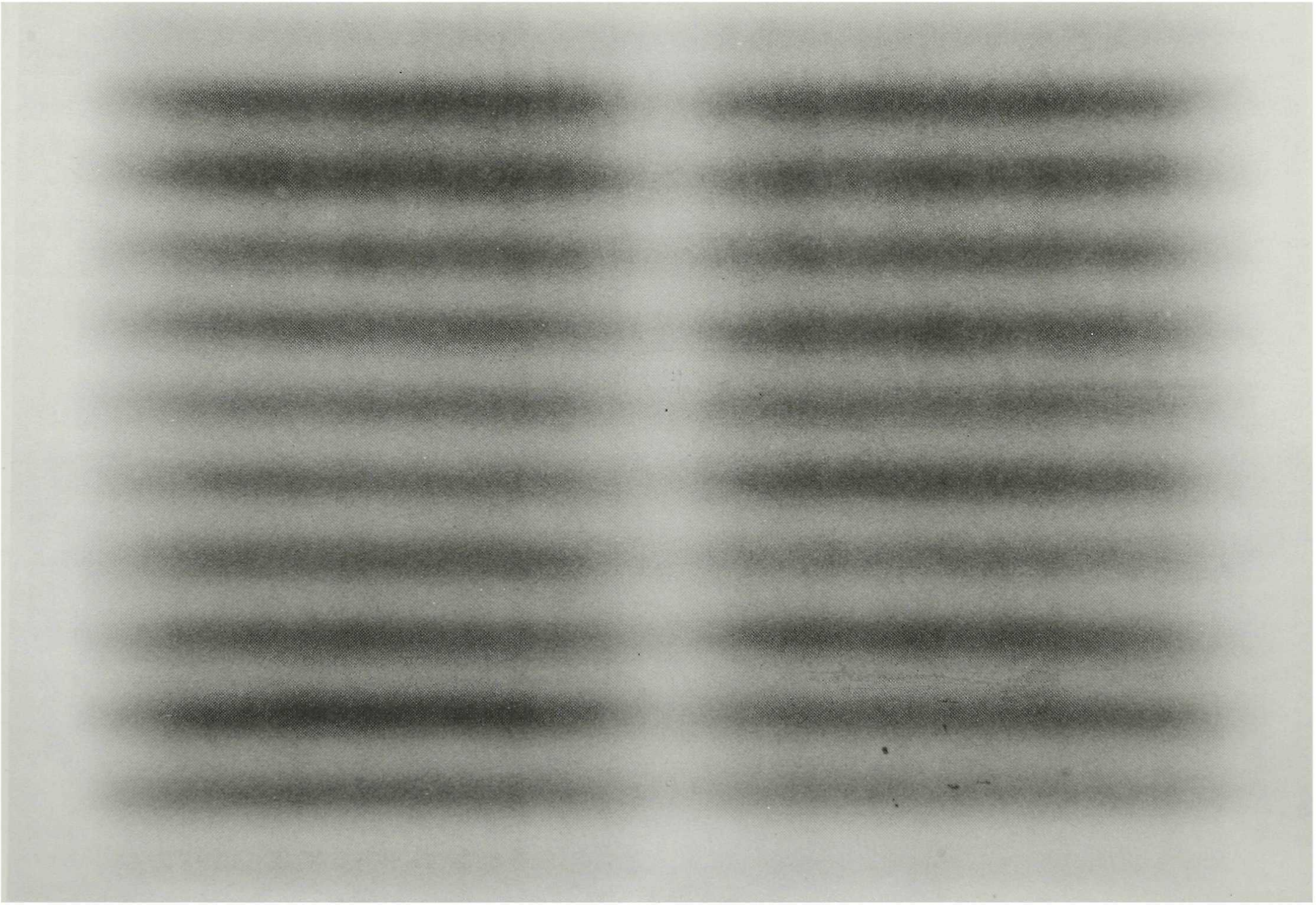

Fig. 5. Autoradiograph of 24 plutonium-containing 4" plates in an Argonne-West canister. ANL Neg. No. 150-77-10.

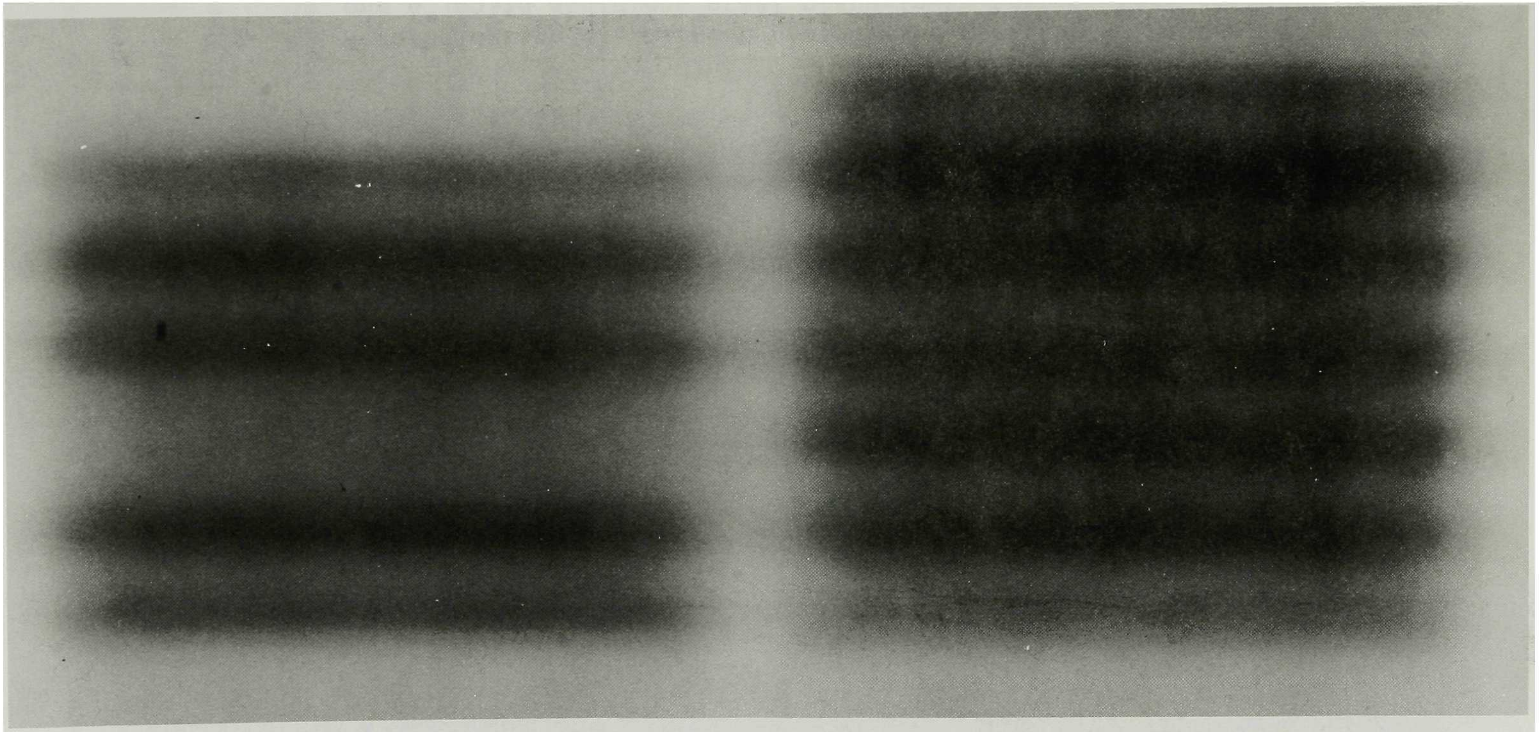

Fig. 6. Autoradiograph of 12 plutonium-containing 4" plates in an Argonne-West canister showing ease of detection of a missing plate. ANL Neg. No. 150-77-2A.

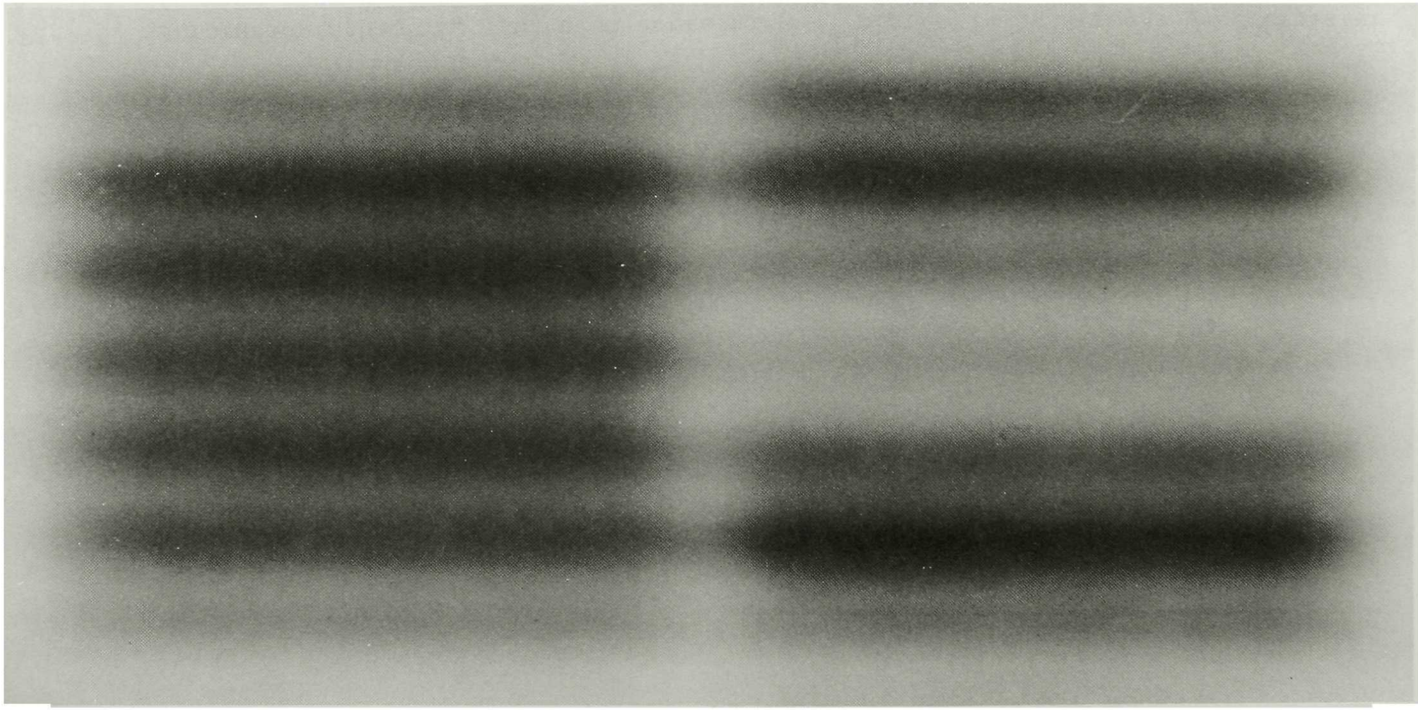

Fig. 7. Autoradiograph of thirteen plutonium-containing 4" plates and one 4" solid stainless steel plate in an Argonne-West canister. ANL Neg. No. 150-77-8.

image of this low-density plate is less dense than that of the steel plate in Fig. 7.

Plate length can also be easily verified. Figure 8 shows a canister with two 4"-long plates in each of the top two rows, a 5" plate in the third row, a 6" plate in the fourth row, and two 4" plates in each of the bottom two rows.

A full-scale inventory of plates contained in about 800 canisters at Argonne-West was conducted with the use of the autoradiographic technique. The inventory was successfully verified, and there was a substantial saving in personnel time and radiation exposure, and in interference with normal facility operations. The technique was not successful at verifying the rather small inventory of 1/8"-thick plates. These thinner plates were sometimes stored in pairs in the canister, and it was not possible to distinguish whether there was one or two plates in a given location.

B. Rods

Some fast critical assembly fuel is in the form of stainless steel-clad rods containing pellets of mixed, plutonium and depleted-uranium oxides. Most of these rods are 6" long, 3/8" in diameter, and have an 0.012"-thick wall. Each rod has an 0.100"-thick end cap welded on one end, and an 0.100"-thick cap plus spacers up to 0.140" thick at the other end.

1. Argonne-East Storage

Fuel rods at the Argonne-Illinois site are stored in cylindrical canisters made of 1/8"-thick aluminum. The canisters are approximately 6.5" in diameter and contain 114 tightly packed aluminum tubes, with the tube axes parallel to the canister axis. Each tube can accommodate one fuel rod. There is 0.110" between the walls of adjacent rods.

Autoradiographs can be obtained of the fuel rods in these

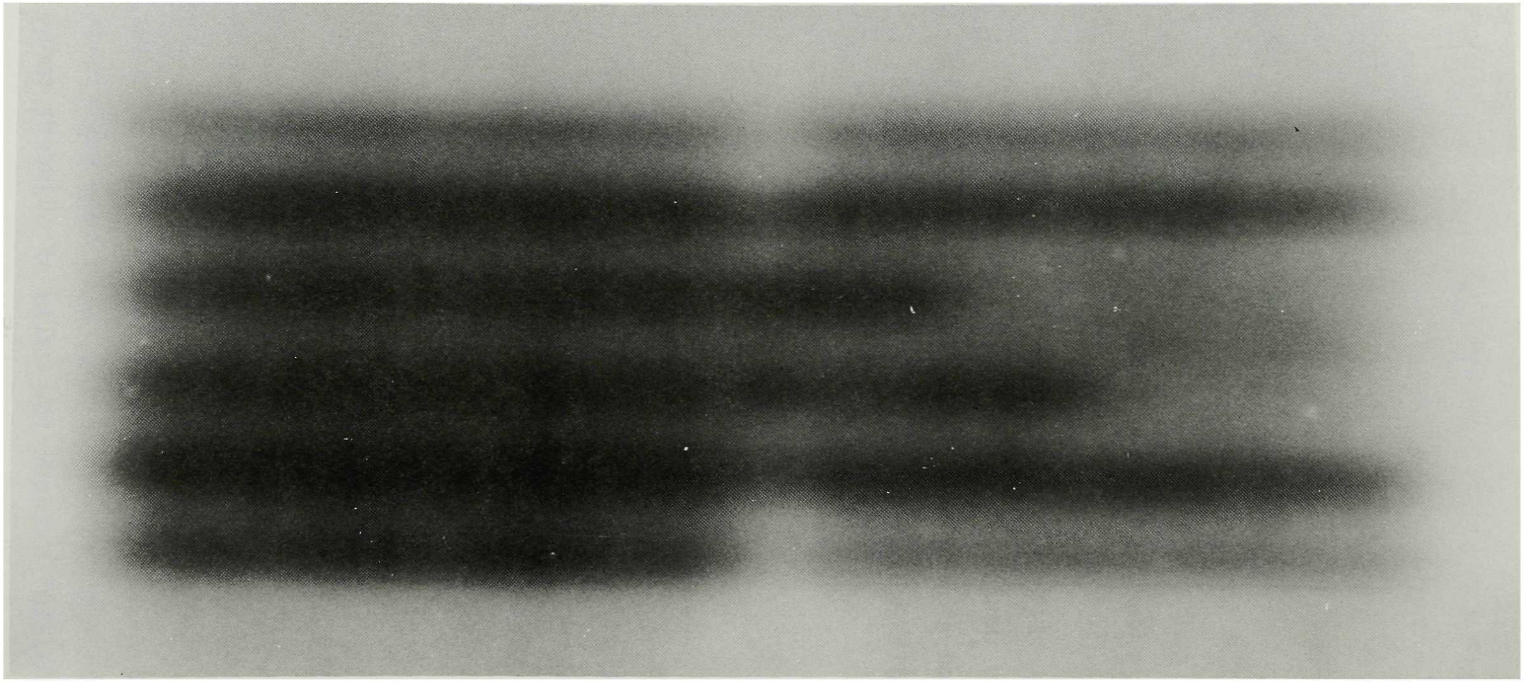

Fig. 8. Autoradiograph of 10 plutonium-containing fuel plates of various lengths. ANL Neg. No. 150-77-9.

canisters by placing a film packet under the flat canister bottom and by exposing the film for approximately 90 minutes. It is necessary to orient all rods with the 0.100"-thick end cap toward the film. The variable-thickness spacers at the other end will result in variable gamma-ray absorption and, consequently, in variable image density for identical fuel rods. Fuel-rod images are sufficiently clear to allow the number of rods to be counted. A missing rod in an array can be detected and is characterized on the film by a region of increased density. This increase in density is due to the intensity of radiation from the rods surrounding the void being greater than the radiation penetrating the end cap of a rod. The substitution of a rod containing depleted uranium can also be detected and is characterized on the film by a region of reduced density. The autoradiographs of rods in Argonne-East storage canisters do not have sufficent contrast to allow photographic reproduction.

Argonne-East rod canisters are stored in "birdcages" which provide minimum separation for criticality considerations. Because of this storage method, it was not possible to place film under the canisters in the vault. Thus the autoradiographic technique was not used in any routine inventory of fuel rods.

2. Argonne-West Storage

Fuel rods at the Argonne-Idaho site are stored in the same type of canister as are the fuel plates. The canisters have been modified by removing the posts which support the plates. The rods are held in an insert which resembles a test-tube rack. It can accommodate 4 rows of 16 rods each. The rods are oriented across the width of the canister. In this storage configuration, inventory by autoradiography has not been possible. Film placed above or below the canisters only gives information about the top or bottom row of rods and gives no information about the interior rods. Film placed along the edge of the canister will show images of the ends of rods, but these images are very diffuse and are difficult to interpret. The images are diffuse because of the rather large film-rod distance (0.6") and because of the poor film-canister contact. The close spacing, 0.050" between rods, also makes the autoradiographs difficult to interpret. Single missing or substituted inert rods cannot be reliably detected. Consequently, autoradiography was not used in the inventory of fuel rods at Argonne-West.

III. FUEL IN THE REACTOR

While it is in operation, one of the fast critical assemblies (zero-power reactors) at Argonne may contain a significant portion of the plutonium fuel inventory. Thus, any inventory verification must include material contained in the assembly itself. Autoradiography has great promise as a technique for conducting an *in situ* assembly inventory verification for plutonium-containing fuel elements with a minimum of inventory time, of radiation exposure to personnel, and of disruption of normal operations.

The Argonne fast critical assemblies consist of pairs of arrays of fuel-containing drawers. Each drawer is 2" x 2" x 36", and is held in place by a steel matrix. Fuel plates are loaded into a drawer on their edges, normally forming one or two 18"-long columns. The remaining space in the drawers is filled with such diluent materials as depleted uranium, sodium, graphite, stainless steel, or aluminum.

An autoradiograph of the plutonium-containing fuel plates in an assembly drawer can be obtained by slipping a prepackaged film strip between the top of the contents of a drawer and the steel matrix, and by exposing the film for approximately 15 minutes. Examples of assembly drawer autoradiographs are shown in Fig. 9. Figure 9-A shows three fuel plates in a single column, arranged end-to-end with the cladding touching. The plate at the top of Fig. 9-A is 8" long; the center and the bottom plates are 5" long, with the bottom plate only partially visible. The sharpness of the plate images and the short exposure times result from the short film-plate distance of about 0.020". Figure 9-B shows a drawer with two columns of fuel. Each column consists of a 7"-, a 6"-, and a 5"-long plate. Figure 9-C shows an autoradiograph of a drawer containing diluent material but no fuel. The drawer directly above the one shown in 9-C contains a double column of fuel. Thus, Fig. 9-C shows that the

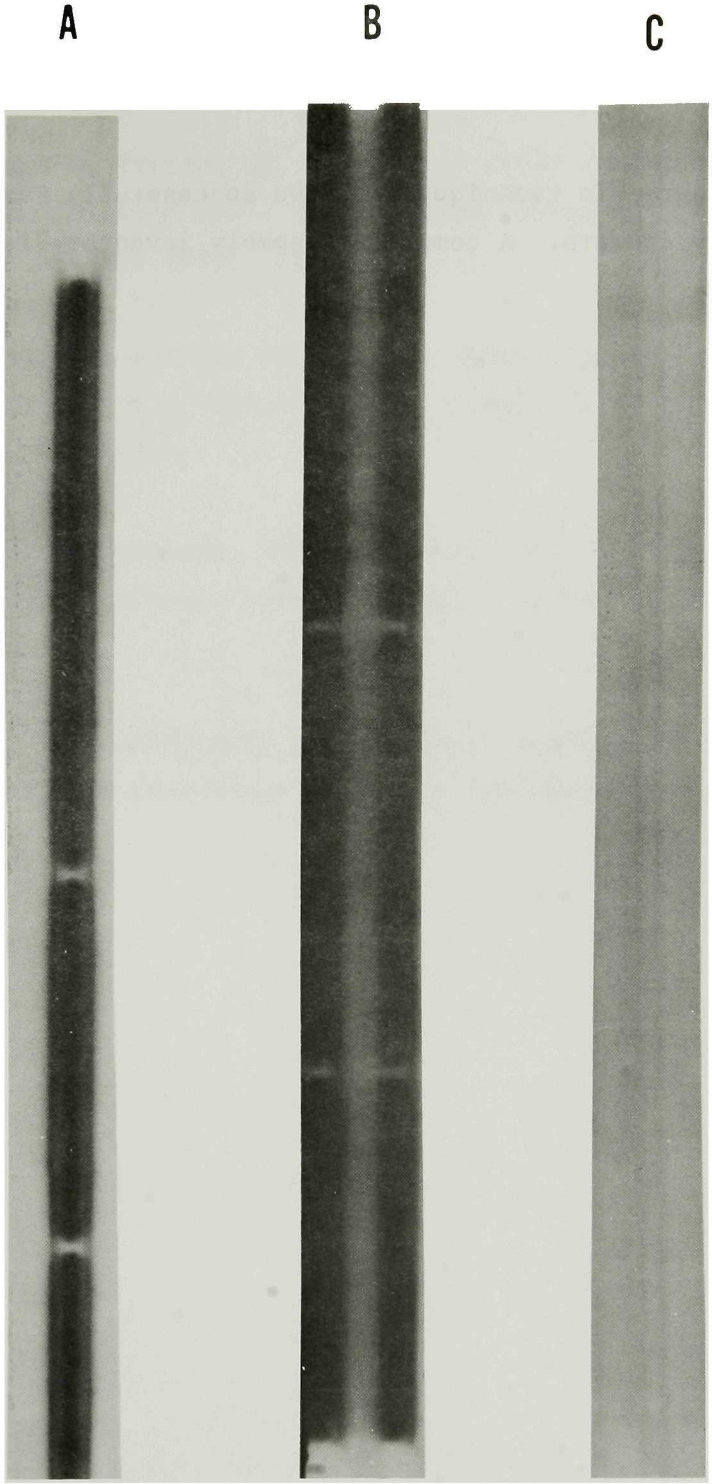

Fig. 9. *In situ* autoradiographs of contents of zero-power reactor drawers. A. Single Column of plutonium-containing fuel plates. B. Double column of plutonium-containing fuel plates. C. Empty drawer with drawer above having double column of plutonium-containing fuel plates. ANL Neg. No. 150-77-4.

0.110"-thick steel between drawers absorbs enough gamma radiation to eliminate contributions from the contents of one drawer to an autoradiograph of the contents of an adjacent drawer.

The autoradiographic technique has been successfully tested on a sample of assembly drawers. A complete assembly inventory is planned.

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